THEORETICAL CALCULATIONS OF RADIANT HEAT TRANSFER PROPERTIES OF PARTICLE-SEEDED GASES

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16. Abstract

A discussion of the mechanisms of radiant heat transfer in gases containing particles is presented. The Mie theory equations for spherical particles were used to calculate the adsorption, extinction, and scattering parameters of carbon at 3590° F, silicon at 80° F, and tungsten at 76° F, 1530° F, and 2420° F over the wavelength range for which complex refractive index data are available. The application of these results to radiant heat transfer calculations for the gaseous core nuclear rocket is discussed.

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FOREWORD

A discussion of the theoretical basis for research regarding the
absorption of thermal radiation by gases containing particles is pre-
sented herein. One of the major applications is to gaseous reactor
concepts for nuclear rocket propulsion. This work was performed under
NASA Grant NGR-11-002-068 with Mr. Charles C. Masser, Nuclear Systems
Division, NASA Lewis Research Center as Technical Manager.

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LIST OF SYMBOLS

a	Radius of a spherical particle.
$\mathring{A}$	Angstroms.
a_n, b_n	Mie coefficients.
c	Velocity of light.
dA	Element of surface area.
$\vec{E}$	Electric component of an electromagnetic wave.
$\vec{H}$	Magnetic component of an electromagnetic wave.
$H_r^{(2)}$	Spherical Hankel function of the second kind.
I	Radiant energy intensity.
j	Emission coefficient.
J_r	Spherical Bessel function.
k_a	Linear absorption coefficient of the particle-seeded gas.
k_a^g	Linear absorption coefficient of the gas.
k_a^P	Linear absorption coefficient of the particles.
k_s	Linear scattering coefficient of the particle-seeded gas.
k_s^P	Linear scattering coefficient of the particles.
k_T	Total linear attenuation coefficient.
n	Integer between 1 and ∞.
N	Relative refractive index of a particle.
p	Scattering amplitude function.
P_n	Legendre polynomial.
q	Size parameter $(= 2\pi a/\lambda)$
q_r	Emitted radiant energy.
R_e	Denotes the real part of a complex number.
s	Distance.
t	Time.
V	Volume of a spherical particle.
x, y, z	Position coordinates.
ϵ	Inductive capacity.
θ	Angle, generally angle of scattering.

λ	Wavelength.
μ	Magnetic permeability.
μ_a	Absorption parameter of the particles.
μ_e	Extinction parameter of the particles.
μ_s	Scattering parameter of the particles.
ρ	Mass of particles per unit volume of particle-seeded gas.
ρ_P	Mass density of the particle material.
σ	Electrical conductivity.
σ_a	Absorption cross section of a spherical particle.
σ_e	Extinction cross sections of a spherical particle.
σ_s	Scattering cross section of a spherical particle.
Ω	Solid angle.
ϖ_n	Coefficient of a Legendre polynomial.

RADIANT HEAT TRANSFER IN

PARTICLE-SEEDED GASES

by J.R. Williams, A.S. Shenoy and J.D. Clement

SUMMARY

The mechanisms of radiant heat transfer in gases containing particles
are examined to provide a theoretical basis for experimental research into
the absorption and scattering characteristics of particle seeded gases.
The Mie theory equations for spherical particles were used to calculate the
absorption, extinction and scattering parameters of submicron-sized particles
of carbon at 3590°F, silicon at 80°F, and tungsten at 76°F, 1530°F and 2420°F
over the wavelength range for which complex refractive index data are avail-
able.

From an examination of the latest available data on the thermal emis-
sion spectrum of hot gaseous uranium it was concluded that the wavelength
range of interest for gaseous reactor heat transfer calculations lies be-
tween 2000 and 8000 Angstroms. Hydrogen at temperatures below about 16,000°F
is quite transparent to thermal radiation in this wavelength range. In this
wavelength range the absorption parameter of particle clouds is greatest for
particles with diameters of 0.1 micron to 0.3 microns. The maximum absorption
parameter of spherical carbon particles is about 40,000 cm^2/gm for particles
of about 0.2 microns in diameter. For tungsten particles of the same size
the absorption parameter is about 5000 cm^2/gm, however, tungsten particles
scatter much more energy than carbon. Silicon particles with diameters of 0.2
microns or less have a higher absorption parameter than either carbon or
tungsten particles of the same size.

INTRODUCTION

At high temperatures thermal radiation may be an important means of transferring heat to a gaseous coolant. The absorption of thermal radiation in a gas is usually greatly enhanced by the addition of very small particles of a suitable material. If the particles are small enough, they may remain essentially in thermal and dynamic equilibrium with the gas as the gas is heated by radiation to the particles with subsequent conduction from the particles to the gas.[1,2] Particle-seeded gases are currently being considered as coolants for gas core reactors which may be used for highly efficient powerplants and rocket propulsion and for other applications such as reentry shielding[3] and rocket nozzle cooling[4] which require that surfaces be protected from intense thermal radiation.

Two gaseous core nuclear reactor concepts that are currently being investigated are the coaxial flow reactor,[5,6] in which a slow moving central stream of gaseous fissioning fuel heats a fast moving annular stream of particle-seeded gas primarily by thermal radiation, and the nuclear light bulb reactor,[7,8] in which fissioning fuel is contained in a transparent partition and a particle-seeded gas is heated by thermal radiation through the partition. The particles in the gas are necessary to insure sufficient radiant heat transfer to the working fluid and to minimize heating of the containment vessel.[9] Possible uses for gaseous reactors include nuclear rocket propulsion, large efficient power plants using MHD conversion,[10] certain industrial processes, and other applications which require extremely high temperatures and power densities. The performance of a gaseous core nuclear rocket would be far superior to any present-day rocket,[11] chemical or nuclear. Combining a MHD generator with a gaseous reactor would more or less completely remove any constraint on the top temperature of the thermodynamic cycle and would result in much more efficient power generating plants.[10] Thermal pollution would thereby be reduced, and also space power systems could operate at much higher radiator temperatures.

Since a gaseous reactor would rely on thermal radiation from the hot fissioning core to heat a gaseous coolant, particle seeding is essential.

to its operation. Whereas gases alone tend to absorb thermal radiation in lines and bands, the absorption characteristics of particle clouds vary only gradually with wavelength.[12-14] Particle clouds of some refractory materials have been shown to have very high absorption coefficients such that only a very small mass fraction of such a particle seed material dispersed in a gas will make the gas highly absorbing.[15-16]

Radiant energy absorption by particle clouds has been investigated for many years by many people. A rigorous solution of Maxwell's equations for the absorption and scattering of radiant energy by spherical particles was published in 1908 by Mie,[17] and since that time a considerable amount of effort has been spent solving the Mie equations under various conditions.[18] Since the gaseous core nuclear rocket would utilize particle-seeded hydrogen as the propellant, work was recently begun at Georgia Tech to measure the absorptive properties of hydrogen seeded with submicron-sized particles at high temperatures and pressures. In order to better understand the experimental data and to make a judicious choice of seed materials, the Mie equations have been solved for some of the seed materials used in these experiments. The following pages present much of what has been learned regarding the theoretical basis of this experimental program. The experimental program itself is described in a separate contractor report.[30]

GENERAL BACKGROUND

There are three significant mechanisms by which radiant energy interacts with a gas containing particles: (1) absorption by the gas, (2) absorption by the particles suspended in the gas, and (3) scattering by the particles. The importance of each of these three mechanisms depends on the composition, temperature and pressure of the gas, the composition, sizes and shapes of the particles, the particle number density, and the spectrum of the radiant energy.

The attenuation of a beam of monochromatic radiant energy by a gas containing particles is governed by the expression

$$I(\lambda,x) = I(\lambda,o) \, e^{-k_T(\lambda)x} \tag{1}$$

where $k_T(\lambda)$ is the total linear attenuation coefficient for radiant energy of wavelength λ, and x is the distance the beam traverses through the seeded gas. The total linear attenuation coefficient for all three interaction processes is equal to the sum of the linear attenuation coefficients for each process separately, that is,

$$k_T(\lambda) = k_a^g(\lambda) + k_a^p(\lambda) + k_s^p(\lambda) \tag{2}$$

where $k_a^g(\lambda)$ is the linear attenuation coefficient due to absorption by the gas alone, $k_a^p(\lambda)$ is the linear attenuation coefficient due to absorption by the particles, and $k_s^p(\lambda)$ is the linear attenuation coefficient due to scattering by the particles. $k_a^p(\lambda)$ and $k_s^p(\lambda)$ are proportional to the number density of the particles as long as the particles are randomly oriented and the average distance between the particles is much greater than their effective radius, so it is convenient to define the absorption parameter $\mu_a(\lambda)$ and scattering parameter $\mu_s(\lambda)$ by

$$\mu_a(\lambda) = \frac{k_a^p(\lambda)}{\rho} \quad \text{and} \quad \mu_s(\lambda) = \frac{k_s^p(\lambda)}{\rho} \tag{3}$$

where ρ is the particle density in grams of particles per cubic centimeter of aerosol. $\mu_a(\lambda)$ is also called the mass absorption coefficient. The totality of processes by which energy is removed from a beam by a particle cloud is called extinction, so the extinction parameter is given by

$$\mu_e(\lambda) = \mu_a(\lambda) + \mu_s(\lambda) \ . \tag{4}$$

The absorption, scattering, and extinction parameters are independent of the concentration of particles.

One may now consider the absorption of radiant energy by particle-seeded gases to be the sum of two independent processes; the absorption by the gas itself and absorption due to the particles in the gas. The absorption coefficient of the gas, $k_a^g(\lambda)$, depends only on the composition, temperature, and pressure of the gas; whereas the absorption and scattering parameters of the particles, $\mu_a(\lambda)$ and $\mu_s(\lambda)$, depend on the composition, sizes, and shapes of the particles. Thus, $k_a^g(\lambda)$ may usually be determined for the pure gas and $\mu_a(\lambda)$ and $\mu_s(\lambda)$ for the particles in any transparent medium and then $k_T(\lambda)$ is calculated for the particle-seeded gas using equations 2 and 3. However, this procedure becomes difficult, if not impossible, when the composition of the gas and the sizes and shapes of the particles are changed by chemical reactions between the particles and the gas.

The basic mechanism of radiant energy absorption by particle clouds and by gases are quite different. Since atoms and molecules of a gas absorb radiant energy in discrete quanta, the absorption coefficient of a gas may change many orders of magnitude over a wavelength interval of a few Angstroms. The familiar absorption spectra of various gases attest to the wide variations of $k_a^g(\lambda)$ as λ is changed.

Whereas gases tend to absorb in lines and bands, the absorption and scattering characteristics of particle clouds vary only gradually with the wavelength of the incident radiant energy. Thus, $\mu_a(\lambda)$ and $\mu_s(\lambda)$ are smoothly varying functions of wavelength. Scattering enhances energy absorption in particle clouds by increasing the average path length traversed by the radiant energy. However, in any given unit volume of aerosol, the particle-gas mixture is heated only by absorption, not by scattering. For this reason it is convenient to define the absorption coefficient for the

aerosol, $k_a(\lambda)$, by

$$k_a(\lambda) = k_a^g(\lambda) + k_a^p(\lambda) = k_a^g(\lambda) + \rho\mu_a(\lambda) \; . \tag{5}$$

Then the scattering coefficient for the aerosol is equal to the scattering coefficient of the particles alone, since scattering by the gas is negligible.

$$k_s(\lambda) = k_s^p(\lambda) = \rho\mu_s(\lambda) \; . \tag{6}$$

The effect of scattering depends not only on the value of $\mu_s(\lambda)$ but also on the angular dependence of the scattered energy. Scattering from small particles is usually highly anisotropic.

A rigorous solution to Maxwell's equations for the absorption and scattering of radiant energy by homogeneous spherical particles of any composition suspended in a homogeneous nonmagnetic transparent medium was published in 1908 by Gustav Mie[17] and is now commonly known as the Mie theory. Krascella[14] applied a transformation procedure developed by Aden[19] to the Mie equations to calculate the effect of particle size, wavelength, and particle temperature on particle opacity in those regions of the ultraviolet, visible, and infrared spectra for which complex index of refraction data were available. The authors have used Krascella's program to extend these calculations to other types of particles and to a broader wavelength range.

Svatos[20] has recently published a solution to Maxwell's equations for extinction by flattened ellipsoids; however, at present there is no theory to accurately predict the absorption and scattering characteristics of irregularly shaped particles. Mie theory predicts that $\mu_a(\lambda)$ is a maximum when the particle radius is of the order of the wavelength λ divided by 2π. Thus, for a given particle seed density ρ, the absorption of thermal radiation in the near infrared, visible, and ultraviolet regions of the spectrum is greatest for submicron-sized particles of diameters in the range of 0.05 to 0.25 micron. Submicron-sized particles of refractory materials are generally highly irregular in shape, so Mie theory can only be used as an approximation to the absorption and scattering characteristics of these particles.

RADIANT HEAT TRANSFER IN PARTICLE SEEDED GASES

The spectral thermal radiation intensity $I(\lambda) \cdot \cos\theta \cdot dA \cdot d\Omega \cdot d\lambda \cdot dt$ is defined as the radiant energy absorbed, emitted, or scattered in the wavelength interval λ to $\lambda + d\lambda$ in a time interval t to $t + dt$ in solid angle $d\Omega$ passing through a unit area normal to the surface dA (see Figure 1). $I(\lambda)$ is in general a function of wavelength λ, of time t, of solid angle Ω, and of position coordinates x, y, z, and thus may be written as

$$I(\lambda) = I(\lambda, x, y, z, \Omega, t) \tag{7}$$
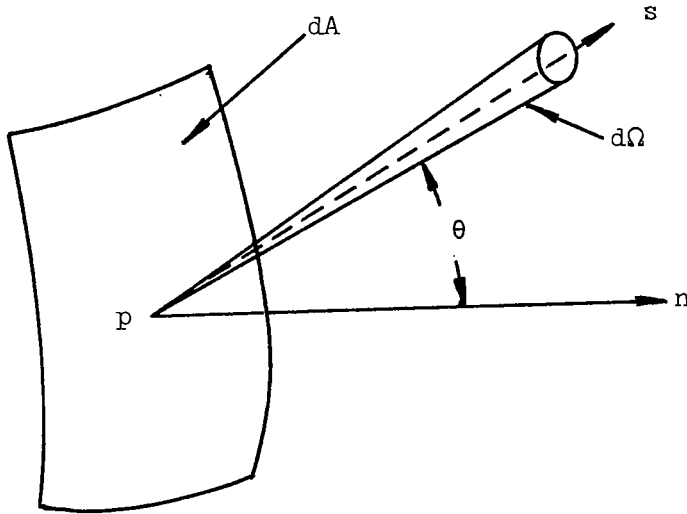
Figure 1. Spectral Radiation Intensity Illustrated

The decrease in $I(\lambda)$ as radiant energy traverses an absorbing medium is given by

$$dI(\lambda) = -k_a(\lambda) \cdot I(\lambda) \cdot ds \tag{8}$$

where $k_a(\lambda)$ is the linear absorption coefficient of the medium and ds represents the distance traversed by the radiant energy as the intensity decreases by $dI(\lambda)$. An absorbing and scattering medium may be characterized by its absorption coefficient $k_a(\lambda)$, its scattering coefficient $k_s(\lambda)$, and its scattering amplitude function $p(\lambda,\cos\theta)$ where θ is the angle of scattering. $k_a(\lambda)$, $k_s(\lambda)$, and $p(\lambda,\cos\theta)$ may be calculated for a cloud of spherical particles in a transparent medium using the Mie theory. For particles with diameters several times the wavelength or larger, practically all of the scattered energy is confined within 10 degrees or less of the original direction of propagation, so $p(\lambda,\cos\theta)$ is large for small values of θ and becomes much smaller for θ greater than about 10 degrees. However, particles with diameters less than about one tenth wavelength exhibit what is called Rayleigh scattering, that is, symmetrical scattering fore and aft. Thus, $p(\lambda,\cos\theta)$ is symmetrical about $\theta = 90°$ and as much energy is backscattered as is forward scattered. The growing tendency towards forward scattering with increasing particle size is known as the Mie effect. In the "Mie region", where the particle diameters are of the same order of magnitude as the wavelength, the scattering coefficient $k_s(\lambda)$ fluctuates widely as the wavelength λ is changed. These fluctuations are due to interference between light transmitted through the particle and the undisturbed beam. There are strong fluctuations of intensity with the angle of observation as well as fluctuations of the total scattering. For absorbing particles these fluctuations are suppressed because the transmitted beam is partially absorbed and the interference is reduced.

In an absorbing and scattering medium, the rate at which energy is scattered by a unit volume of scattering medium from a beam of intensity $I(\lambda,\Omega)$ from the solid angle $d\Omega$ into the solid angle $d\Omega'$ is

$$k_s(\lambda)I(\lambda,\Omega)p(\lambda,\cos\theta)\frac{d\Omega'}{4\pi}d\lambda d\Omega \qquad (9)$$

where θ is the angle of scattering. The rate of loss of radiant energy from the beam due to scattering into all directions is then

$$k_s(\lambda)I(\lambda,\Omega)d\lambda d\Omega \int_0^{4\pi} p(\lambda,\cos\theta)\frac{d\Omega'}{4\pi} \qquad (10)$$

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Usually it is convenient to normalize the amplitude function to unity, so

$$\int_0^{4\pi} p(\lambda, \cos\theta)\frac{d\Omega'}{4\pi} = 1 \qquad (11)$$

For small particles that exhibit Rayleigh scattering the Rayleigh amplitude function may be used

$$p(\lambda, \cos\theta) = \frac{3}{4}(1 + \cos^2\theta) \qquad (12)$$

It may be written in terms of the Mie coefficients if the particle diameters are of the same order of magnitude as the wavelength. The amplitude function may also be expanded in a Legendre polynomial series[21-23] as

$$p(\lambda, \cos\theta) = \sum_{n=1}^{\infty} \overline{w}_n(\lambda)P_n(\cos\theta) \qquad (13)$$

In order to arrive at an equation describing radiant heat transfer in an isotropic medium which absorbs, emits, and scatters thermal radiation, it is necessary to take into account all processes of energy transport in the medium. These are: (1) scattering of incident radiant energy from $d\Omega$ to other angles (loss), (2) absorption of incident radiant energy from $d\Omega$ (loss), (3) scattering of incident radiant energy from other angles into $d\Omega$ (gain), and (4) the emission of radiant energy by the medium into $d\Omega$ (gain). The sum of these four energy changes is then equated to the net change in radiant energy crossing a small volume element of cross section dA and thickness ds.

First using a normalized amplitude function $p(\lambda, \cos\theta)$, the energy lost from $d\Omega$ due to scattering into all other directions is

$$k_s(\lambda)I(\lambda, \Omega)d\lambda d\Omega \qquad (14)$$

Similarly, the rate of energy loss by absorption is

$$k_a(\lambda)I(\lambda, \Omega)d\lambda d\Omega \qquad (15)$$

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The radiant energy scattered into $d\Omega$ from other directions is

$$k_s(\lambda)d\lambda d\Omega \int_0^{4\pi} I(\lambda,\Omega')p(\lambda,\cos\theta)\frac{d\Omega'}{4\pi} \qquad (16)$$

The energy emitted into $d\Omega$ is

$$j(\lambda)\rho d\lambda d\Omega \qquad (17)$$

where $j(\lambda)$ is the emission coefficient defined by

$$j(\lambda) = \frac{dq_r(\lambda)}{\rho d\Omega d\lambda} \qquad (18)$$

where $dq_r(\lambda)$ is the radiant energy emitted by a unit volume containing a mass density ρ of emitting material into the solid angle $d\Omega$ in the wavelength interval $d\lambda$. In general $j(\lambda)$ depends on the wavelength, position, gas composition, temperature, and time, but not on particle concentration.

For steady state conditions, the sum of these four energy changes is equal to $\frac{dI(\lambda,\Omega)}{ds} d\lambda d\Omega$, so the steady state radiant heat transfer equation is

$$\frac{dI(\lambda,\Omega)}{ds} = -(k_a(\lambda)+k_s(\lambda))I(\lambda,\Omega) + j(\lambda)\rho$$

$$+ k_s(\lambda)\int_0^{4\pi} I(\lambda,\Omega')p(\lambda,\cos\theta)\frac{d\Omega'}{4\pi} \qquad (19)$$

This is the equation for steady state radiant heat transfer in an absorbing, emitting, and scattering medium. It is applicable to particle-seeded gases even if the gas itself absorbs some of the thermal radiation. If the gas is transparent, then the equation may be further simplified by dividing by the particle seed density ρ.

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$$\frac{1}{\rho} \frac{dI(\lambda,\Omega)}{ds} = - \mu_e(\lambda)I(\lambda,\Omega) + j(\lambda)$$

$$+ \mu_s(\lambda) \int_0^{4\pi} I(\lambda,\Omega')p(\lambda,\cos\theta)\frac{d\Omega'}{4\pi} \qquad (20)$$

This is the equation which may be used to evaluate steady state heat transfer through particle-seeded gases. In the time-dependent case, one must take into account enthalpy changes in the aerosol as well as the temperature dependence, and hence time dependence, of the particles and gas.

It is seen from the steady state equation that $\mu_e(\lambda)$, $\mu_s(\lambda)$, and $p(\lambda,\cos\theta)$ are important parameters that must be known over the applicable wavelength range before one may evaluate radiant heat transfer through particle-seeded gases.

SPECTRA OF THERMAL RADIATION SOURCES

Since the absorption and scattering characteristics of particle-seeded gases are wavelength dependent, the spectrum of the thermal radiation is an important parameter for heat transfer calculations. Figure 2 illustrates the spectra of thermal radiation from tungsten at 5000°F, a xenon arc lamp,[15] a uranium-argon plasma,[26] and a uranium plasma.[27] The uranium-argon plasma data were integrated over 100 Å intervals and the uranium plasma data over 500 Å intervals. For this reason, the spectral intensity from the uranium plasma is presented as a histogram.

The uranium plasma and uranium-argon plasma data indicate that the wavelength range of interest for the gaseous core reactor is between 2000 and 8000 Å.
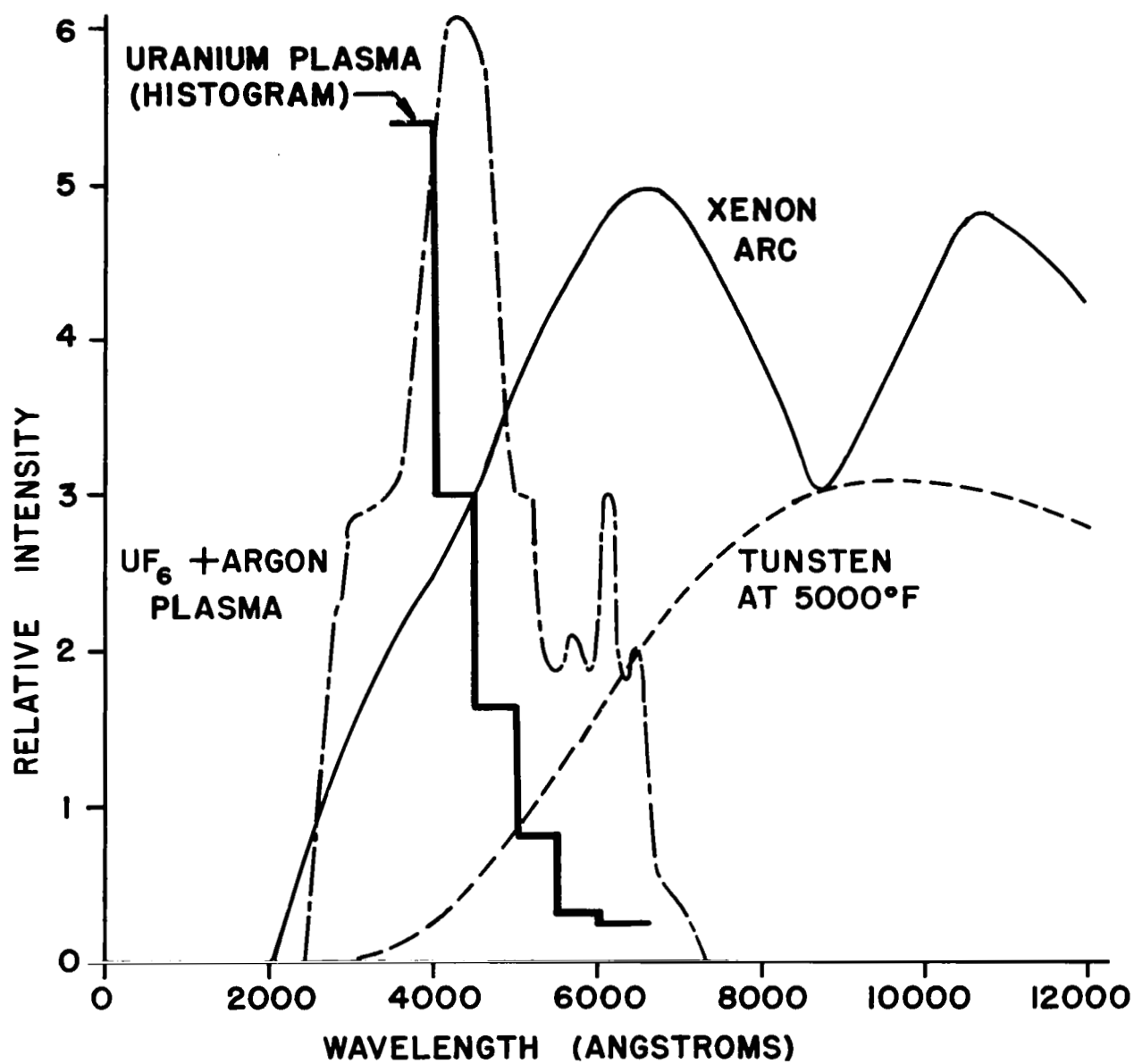
Figure 2. Spectra of Some Intense Thermal Radiation Sources.

RADIANT ENERGY ABSORPTION BY GASES

The mechanism of energy absorption by gases is quite different from that of particle clouds in that individual atoms and molecules absorb energy in discrete quanta. As a result, the absorption coefficient $k_a^g(\lambda)$ may be a strong function of wavelength.

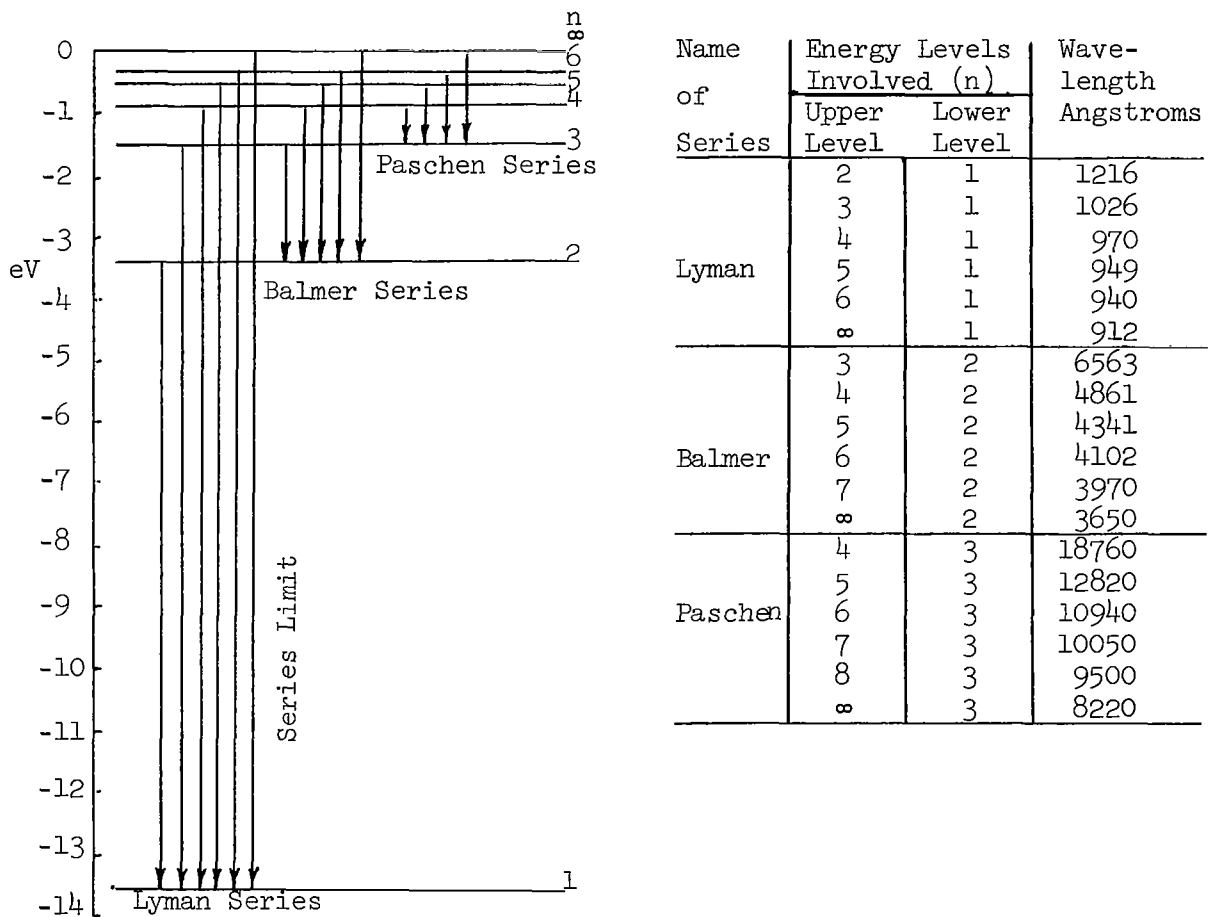The easiest gas to consider in some detail is hydrogen. The energy level diagram of the hydrogen atom is presented in Figure 3.

Name of Series	Energy Levels Involved (n)		Wave-length Angstroms
	Upper Level	Lower Level	
Lyman	2	1	1216
	3	1	1026
	4	1	970
	5	1	949
	6	1	940
	∞	1	912
Balmer	3	2	6563
	4	2	4861
	5	2	4341
	6	2	4102
	7	2	3970
	∞	2	3650
Paschen	4	3	18760
	5	3	12820
	6	3	10940
	7	3	10050
	8	3	9500
	∞	3	8220

Figure 3. Energy Level Diagram and Spectra of the Hydrogen Atom

At low temperatures, the hydrogen atoms are essentially all in the ground state, and radiant energy of 1216 Å wavelength or shorter is required to excite or ionize the atom. This means that hydrogen atoms in the ground state are incapable of absorbing radiant energy of wavelengths greater than 1216 Å. Since the wavelength region of interest for the gaseous core nuclear rocket is above 2000 Å, hydrogen alone at low temperature is seen to be transparent to the thermal radiation from the core. At higher temperatures, however, a significant number of hydrogen atoms are thermally excited to the first excited state (n=2). As is seen from Figure 3, radiant energy of wavelengths as long as 6563 Å will cause ionization. At slightly higher temperatures, the third excited state becomes sufficiently populated to permit significant absorption of radiant energy of wavelengths as long as 18,760 Å. Krascella[28] has calculated the composition, opacity, and thermodynamic properties of hydrogen at high temperatures and pressures. The data for figures 4 and 5 were taken from reference 28 and illustrate the increase in the absorptivity of hydrogen in the wavelength range from 2000 to 8000 Å.

The absorption coefficient, $k_a^g(\lambda)$, of hydrogen at 100 atmospheres pressure from 2000 to 8000 Å is presented in Figure 4 to illustrate the effect of temperature. This effect can be more readily understood by examining Figure 5 which presents the reciprocal of the absorption coefficient, which is equal to the distance over which the radiant energy is attenuated by a factor of e in hydrogen at 100 atmospheres pressure. It is seen that, at temperatures above about 13,000°R, most of the energy would be absorbed in one meter of hydrogen over the whole wavelength range of interest. Since the hydrogen opacity is roughly proportional to pressure, most of the energy would be absorbed over a one meter distance at 10,000°R and 500 atmospheres pressure.
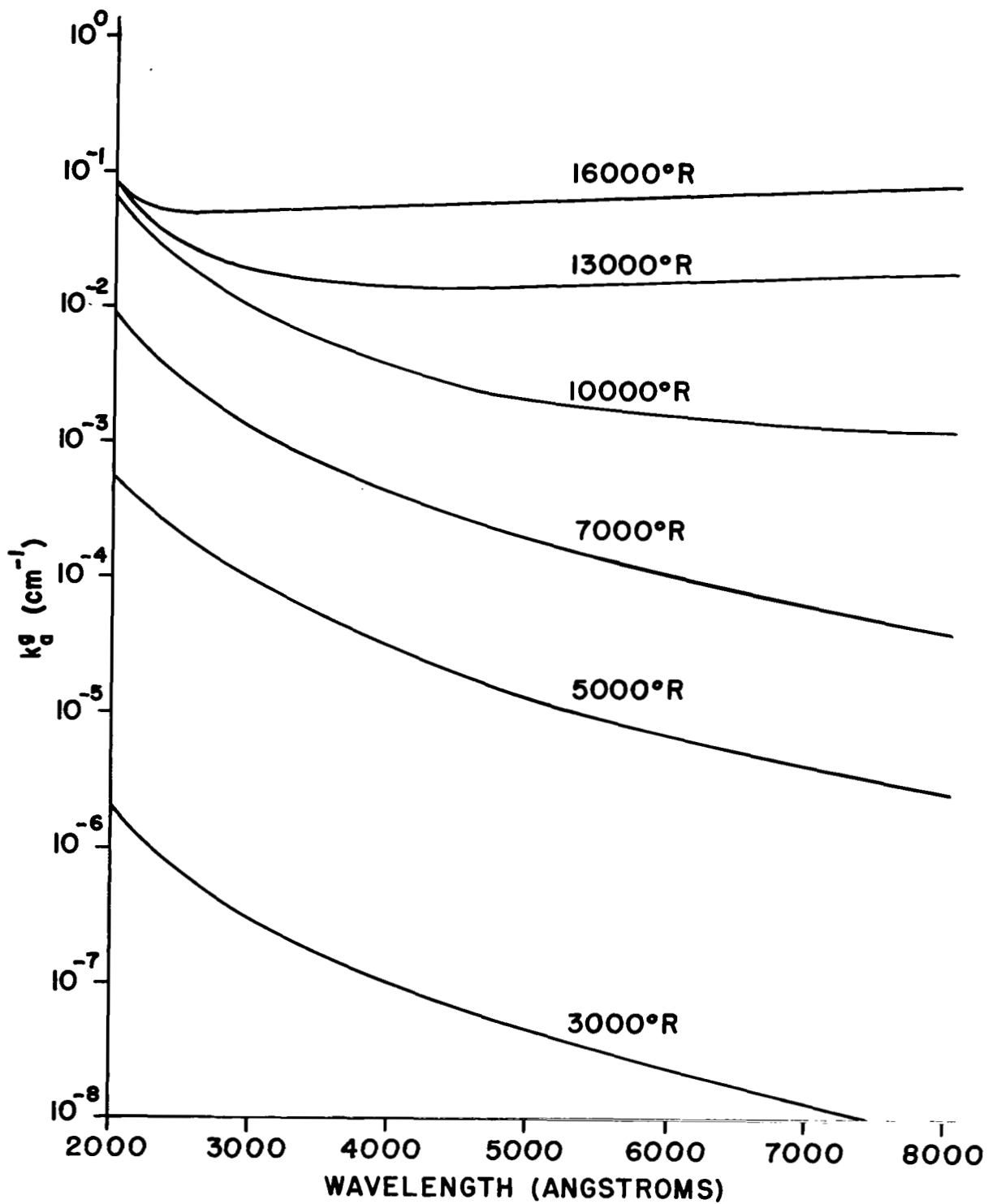
Figure 4.   Absorption Coefficient of Pure Hydrogen at 100 Atmospheres Pressure.
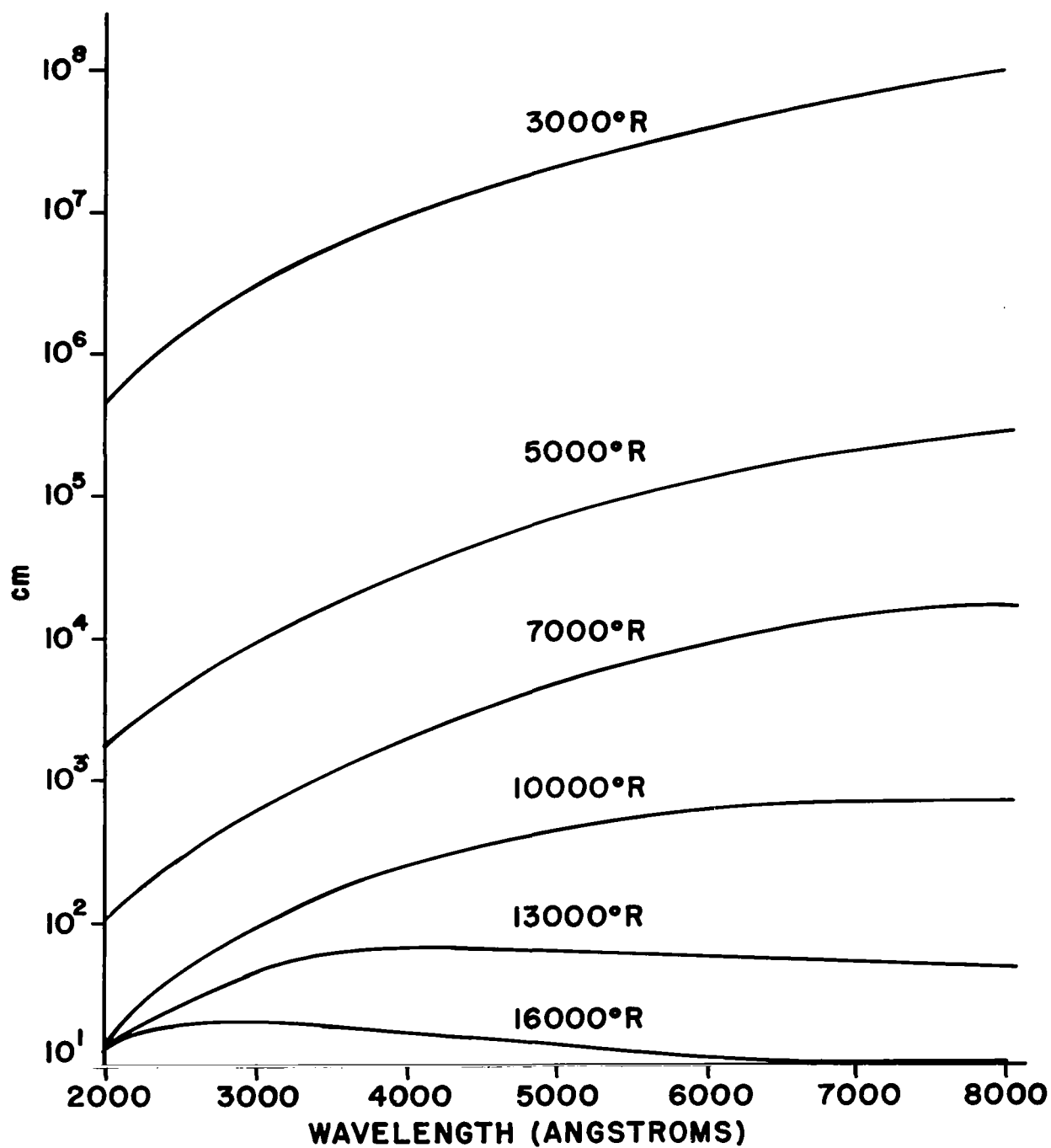
Figure 5. Distance Over Which Radiant Energy Attenuated by e in
Hydrogen at 100 Atmospheres Pressure.

MIE THEORY CALCULATIONS

The Mie theory is the most well known and most useful theory which describes the absorption and scattering of electromagnetic radiant energy by particles. It applies to homogeneous spherical particles of any diameter situated in a homogeneous transparent nonmagnetic medium. Mie solved Maxwell's equations with the appropriate boundary conditions and evaluated the total scattered energy as well as the total energy removed from an incident beam, thus arriving at the scattering and extinction cross sections, σ_s and σ_e. The absorption cross section of the spherical particle is then $\sigma_a = \sigma_e - \sigma_s$, and the extinction, absorption, and scattering parameters are given by

$$\mu_e = \frac{\sigma_e}{\rho_P V} \; , \quad \mu_a = \frac{\sigma_a}{\rho_P V} \; , \quad \text{and} \quad \mu_s = \frac{\sigma_s}{\rho_P V} \tag{21}$$

where ρ_P is the mass density of the material of which the particle is composed and V is the volume of the spherical particle.

Mie's solution, though derived for a single sphere, also applies to absorption and scattering by any number of spheres, provided that they are all of the same diameter and composition and provided also that they are randomly distributed and separated from each other by distances that are large compared to the particle radius. Under these circumstances there are no coherent phase relationships between the light that is scattered by the different spheres, and the total scattered energy is equal to the energy scattered by one sphere multiplied by their total number. Similarly, for a distribution of sizes, the energy scattered by the spheres of each particular size may be summed to obtain the total scattered energy.

In the derivation of Mie's equations, the solution of Maxwell's equations is found which describes the field arising from a plane monochromatic wave incident upon a spherical surface across which the properties of the medium change abruptly. The incident wave and boundary conditions are described in terms of a spherical polar coordinate system centered on the sphere, and the field represented as the sum of two subfields, each of

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which satisfies Maxwell's equations and the boundary conditions. Maxwell's equations together with the boundary conditions then separate into a set of ordinary differential equations which are solved for the two subfields in the form of infinite series. The sum of the solutions for the two subfields is the solution for the actual field. The solution for the scattered wave is used to calculate the desired cross sections for the spherical particles.

Consider the diffraction of a plane, linearly polarized, monochromatic wave by a sphere of radius a immersed in a homogeneous, isotropic medium. We assume the medium to be a non-conductor and both medium and sphere non-magnetic.
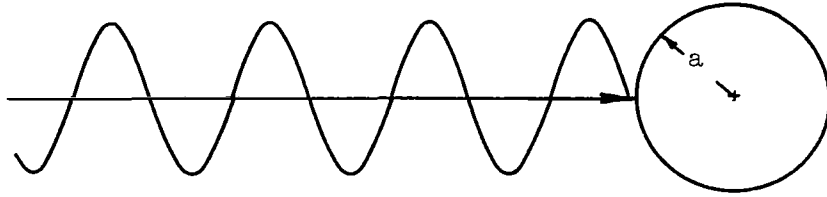
The time-dependent Maxwell's equations with no sources are given by

$$\nabla \times \vec{H}(t) = \frac{4\pi\sigma}{c} \vec{E}(t) + \frac{\epsilon}{c} \frac{\partial \vec{E}(t)}{\partial t} \qquad (22)$$

and

$$\nabla \times \vec{E}(t) = - \frac{\mu}{c} \frac{\partial \vec{H}(t)}{\partial t} \qquad (23)$$

$$\nabla \cdot \vec{E}(t) = 0 \qquad (24)$$

$$\nabla \cdot \vec{H}(t) = 0 \qquad (25)$$

where $\vec{H}(t)$ = magnetic component of the electromagnetic wave

$\vec{E}(t)$ = electric component of the electromagnetic wave

σ = electrical conductivity

ϵ = inductive capacity

μ = magnetic permeability

c = velocity of light

These equations are applicable both inside and outside the sphere. The solution of these equations should satisfy the boundary conditions that the radial and tangential components of the magnetic and electric fields be continuous at the surface of the sphere at $r = a$. In practice, one is usually concerned with scattering of unpolarized light. In such cases, the solutions are modified by averaging over all directions of polarization.

Maxwell's equations together with the boundary conditions separate into a set of ordinary differential equations which are solved to give the Mie equations. Since excellent treatments of Mie's solution are available in the literature,[12,13] a complete derivation is not given here. The basic Mie equations for the extinction and scattering cross sections are given by

$$\sigma_e = \frac{2\pi a^2}{q^2} \cdot \sum_{n=1}^{\infty} (2n + 1) R_e (a_n + b_n) \tag{26}$$

$$\sigma_s = \frac{2\pi a^2}{q^2} \cdot \sum_{n=1}^{\infty} (2n + 1) (|a_n|^2 + |b_n|^2) \tag{27}$$

The symbol "R_e" denotes the real part of a complex number and

a = particle radius

q = size parameter = $\frac{2\pi a}{\lambda}$

λ = wavelength of the electromagnetic wave

and a_n and b_n are the Mie coefficients given by

$$a_n = -\frac{J_n(Nq)\cdot[qJ_n(q)]' - J_n(q)\cdot[Nq\cdot J_n(Nq)]'}{J_n(Nq)\cdot[q\cdot H_n^{(2)}(q)]' - H_n^{(2)}(q)\cdot[NqJ_n(Nq)]'} \qquad (28)$$

$$b_n = -\frac{J_n(q)\cdot[Nq\cdot J_n(Nq)]' - N^2\cdot J_n(Nq)\cdot[q\cdot J_n(q)]'}{H_n^{(2)}(q)\cdot[Nq\cdot J_n(Nq)]' - N^2\cdot J_n(Nq)\cdot[q\cdot H_n^{(2)}(q)]'} \qquad (29)$$

where primes denote differentiation and

$J_r(\ )$ = spherical Bessel function

$H_r^{(2)}(\ )$ = spherical Hankel function of the second kind

$N = \dfrac{\text{refractive index of particle}}{\text{refractive index of medium}}$

The absorption cross section may then be calculated using

$$\sigma_a = \sigma_e - \sigma_s \qquad (30)$$

The above set of equations represent a summary of Mie's formal solution. It is somewhat difficult to calculate the cross sections using these equations. The complications arise due to the relative refractive index N being a complex quantity and also because extensive tables of spherical Bessel functions with complex arguments are not available. An excellent transformation procedure, facilitating computation, is given by Aden[19] which makes possible the calculation of the absorption and scattering coefficients. Krascella[14] has used this transformation of the Mie equations to develop a computer program which calculates σ_a, σ_e, and σ_s in cm^2/particle as a function of material, wavelength of radiation, and particle radius.

This program was obtained from United Aircraft Corporation and the necessary changes made to run it on the UNIVAC 1108 at Georgia Tech. Reference 14 describes this program. The mass density of the particle material is also provided as part of the program to allow the evaluation of the absorption, extinction and scattering parameters using equation 21.

The main inputs to this program are the radius of the particle, wavelength of radiation, temperature, density of the particle material, and the real and imaginary part of the refractive index of the particle material

at each temperature and radiation wavelength.

The calculation of the absorption coefficient is limited by the availability of refractive index data[24], [25] of different particle materials at different temperatures and radiation wavelengths. For some cases where refractive index data were available, the absorption coefficient was calculated by Krascella for various materials at a number of particle radii and radiation wavelengths.

In order to compare new experimental data with the Mie theory, the absorption, scattering, and extinction parameters of carbon, tungsten and silicon were calculated for particle radii from 0.01 micron to 1.0 micron and for radiation wavelengths from 1000 Å to 6000 Å. At lower wavelengths, where refractive index data are not available, the extrapolated values were used. The absorption coefficient was calculated for carbon at 3590°F, silicon at 80°F, and tungsten at 76°F, 1520°F, and 2420°F.

In Figure 6 the absorption coefficient of spherical particles of carbon at 3590°F is presented as a function of radiation wavelength. At lower wavelengths, the absorption coefficient increases with wavelength and for wavelengths above 2800 Å the absorption coefficient is essentially independent of wavelength. Also, one can see that the absorption coefficient increases as the particle size decreases. This effect is more readily seen from Figure 7 which illustrates the absorption coefficient of carbon at 3590°F plotted as a function of particle radius for radiation wavelengths of 2000 Å, 4000 Å, and 6000 Å. As the particle size decreases, the absorption coefficient increases to a maximum value and then decreases with particle size. Figures 8 and 9 illustrate the extinction, absorption, and scattering parameters of carbon. It is seen from figure 8 that scattering may generally be neglected if the diameter is less than the wavelength divided by π.

In Figures 10 and 11, the absorption and scattering coefficients of spherical particles of tungsten are plotted as a function of radiation wavelength at temperatures of 76°F and 2420°F. As can be seen from equations (26) and (27), the absorption and extinction coefficients are not directly a function of temperature, but change with temperature due to .changes in the properties of the seed material, such as the electrical conductivity, inductive capacity, and magnetic permeability. The absorption
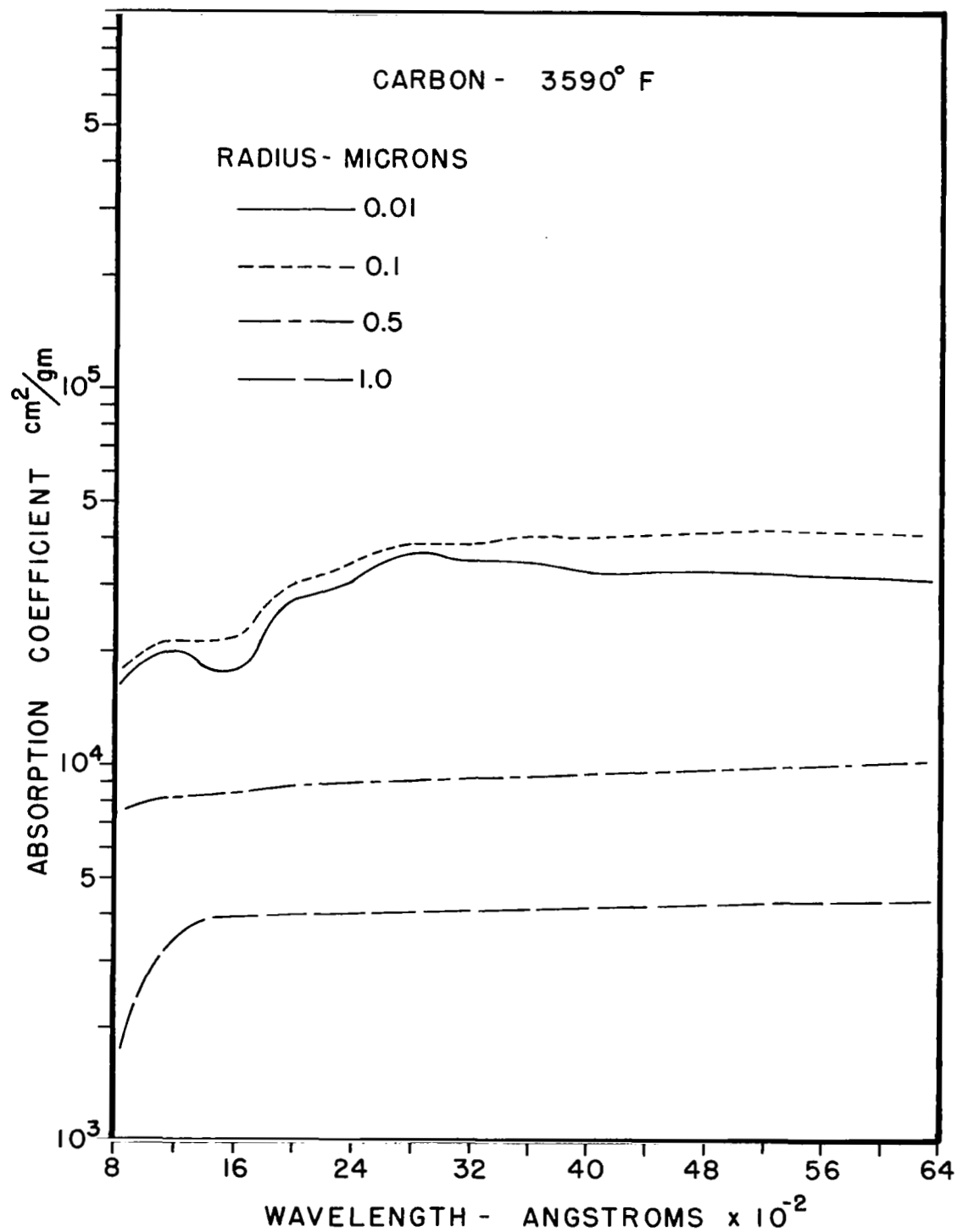
Figure 6.   Theoretical Absorption Coefficient of Carbon as a Function of Radiation Wavelength
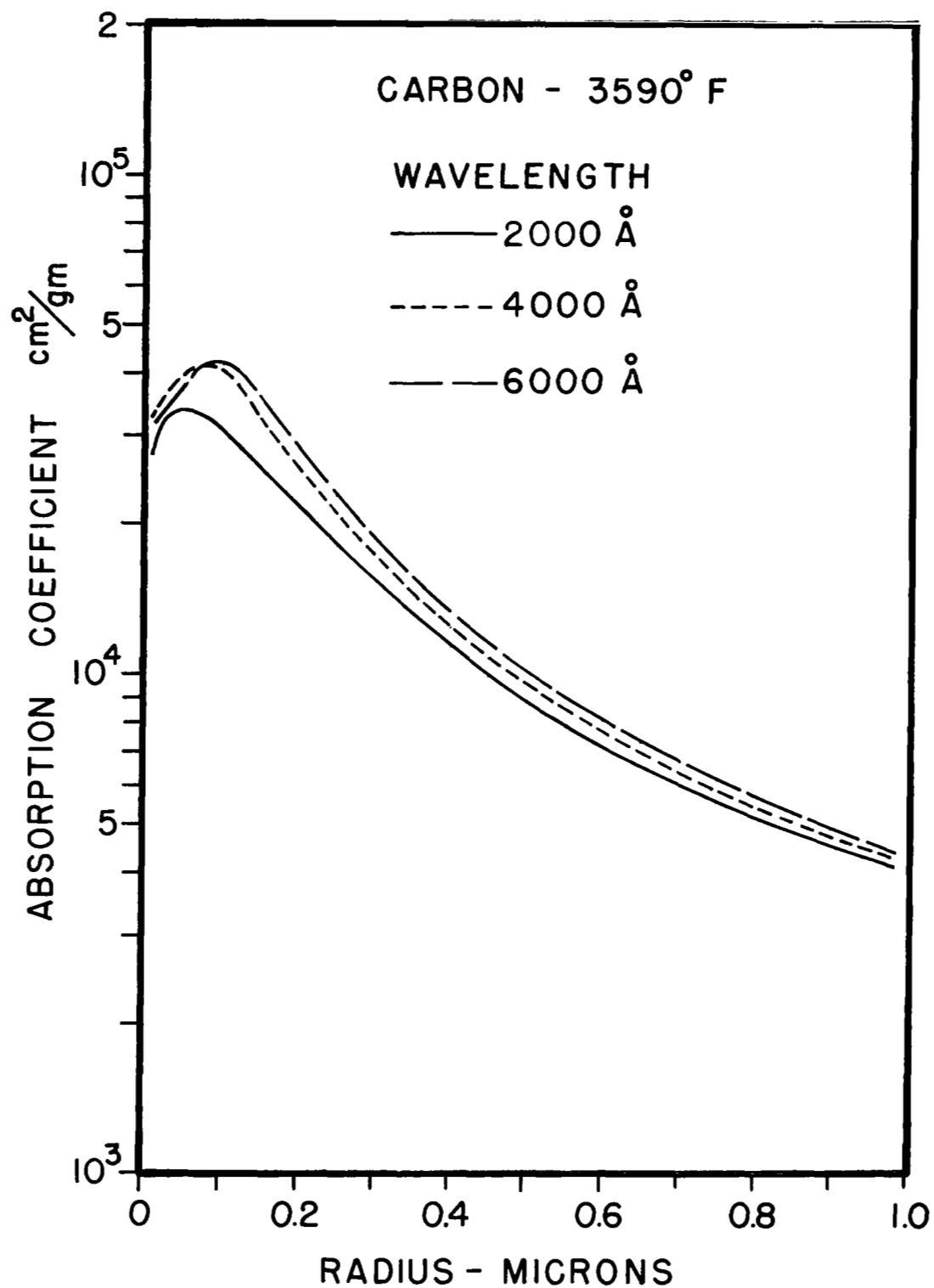
Figure 7.   Theoretical Absorption Coefficient of Carbon as a Function
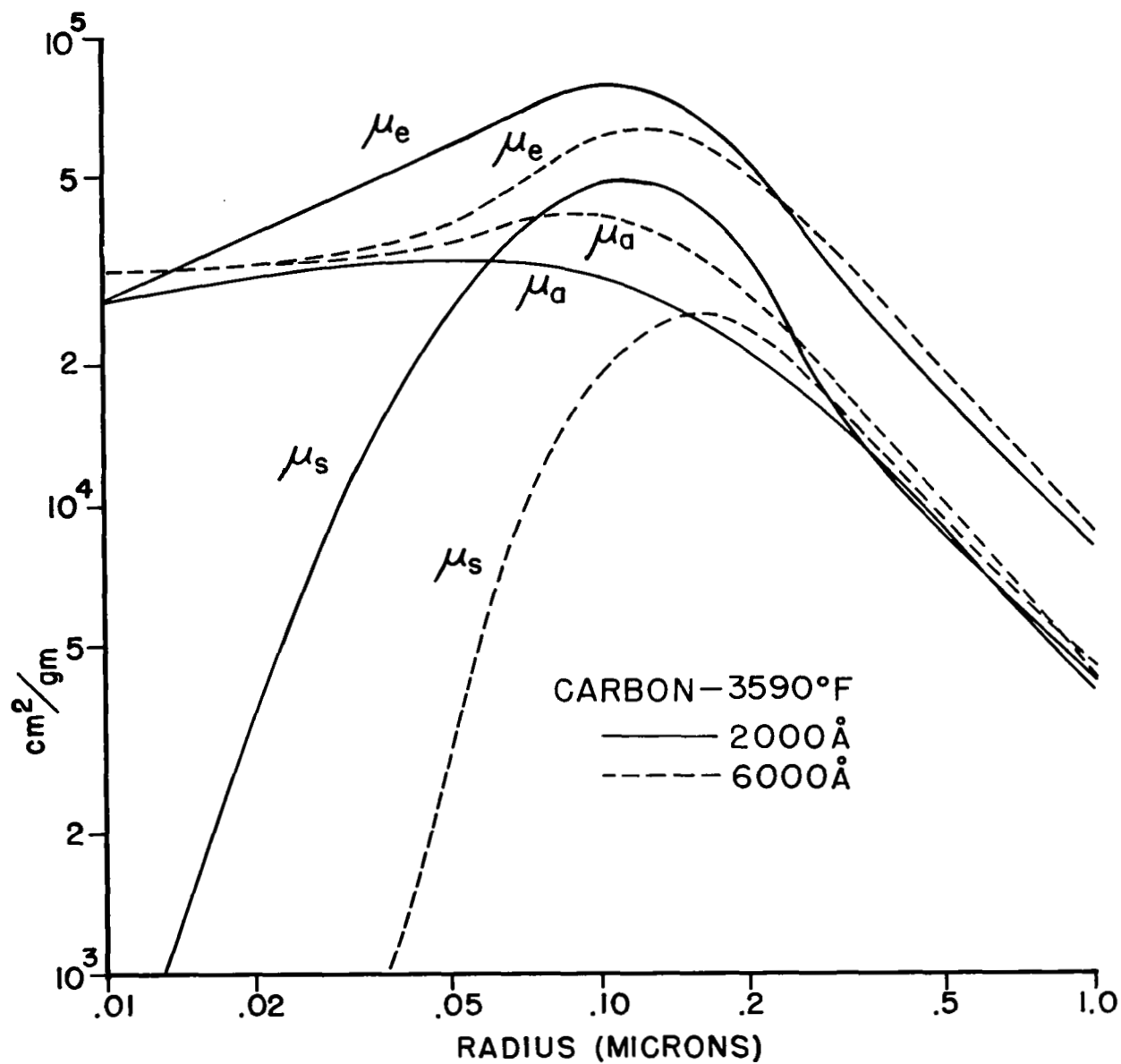of Particle Radius

Figure 8.   Theoretical Absorption, Scattering and Extinction Parameters
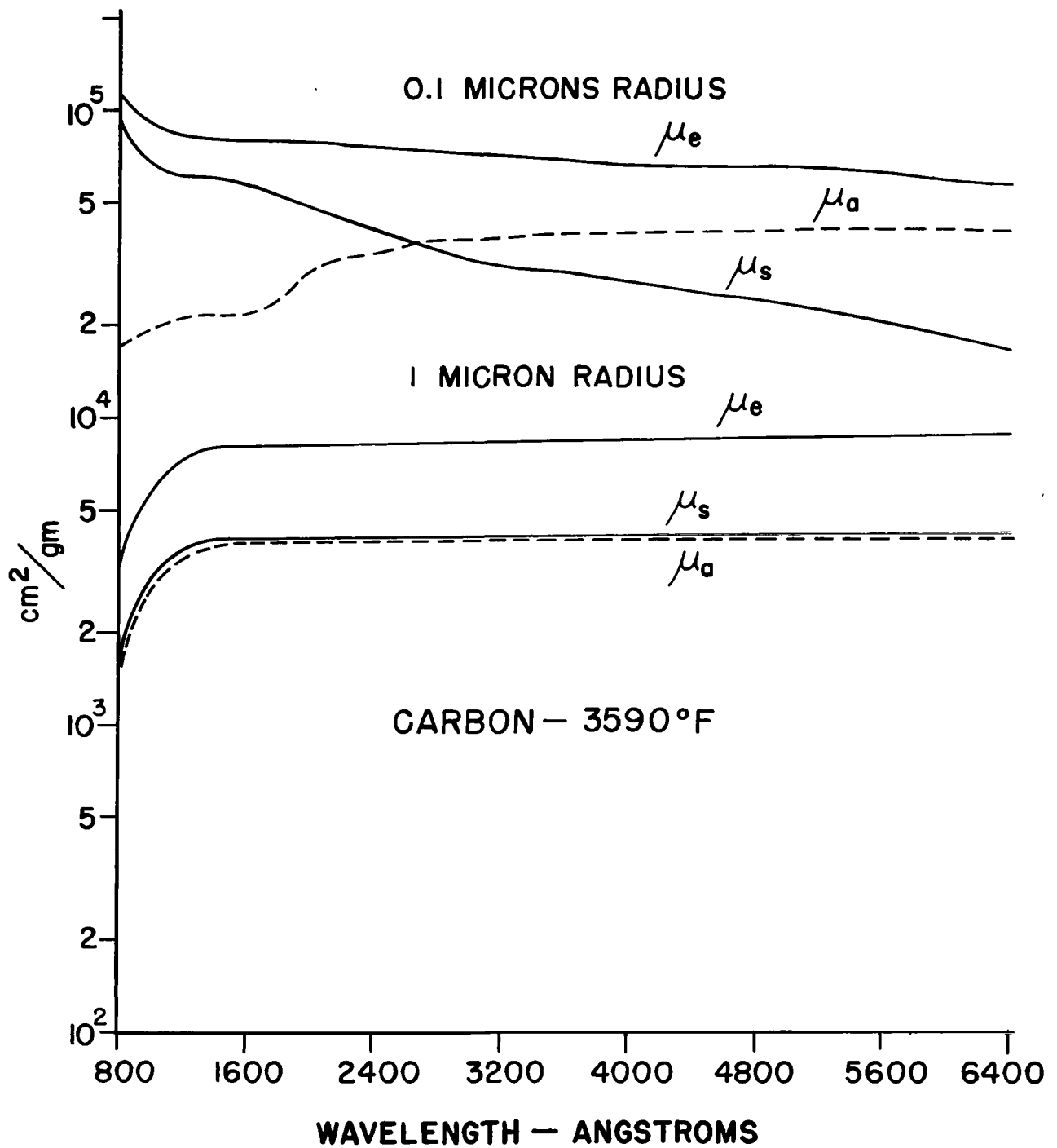of Spherical Carbon Particles.

Figure 9. Theoretical Absorption, Scattering and Extinction Parameters of Spherical Carbon Particles.
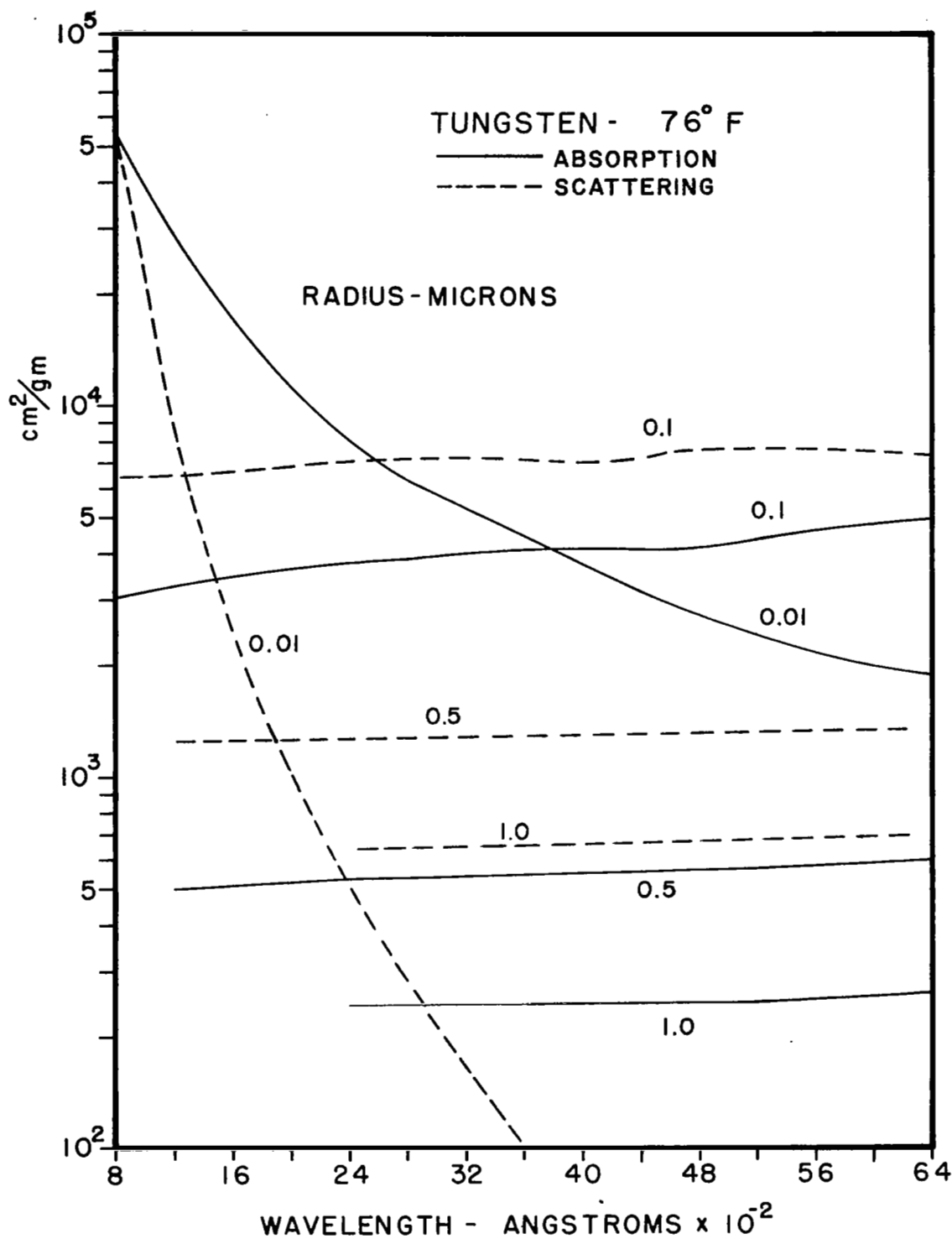
Figure 10. Theoretical Absorption and Scattering Parameters of Spherical Tungsten Particles at 76°F.
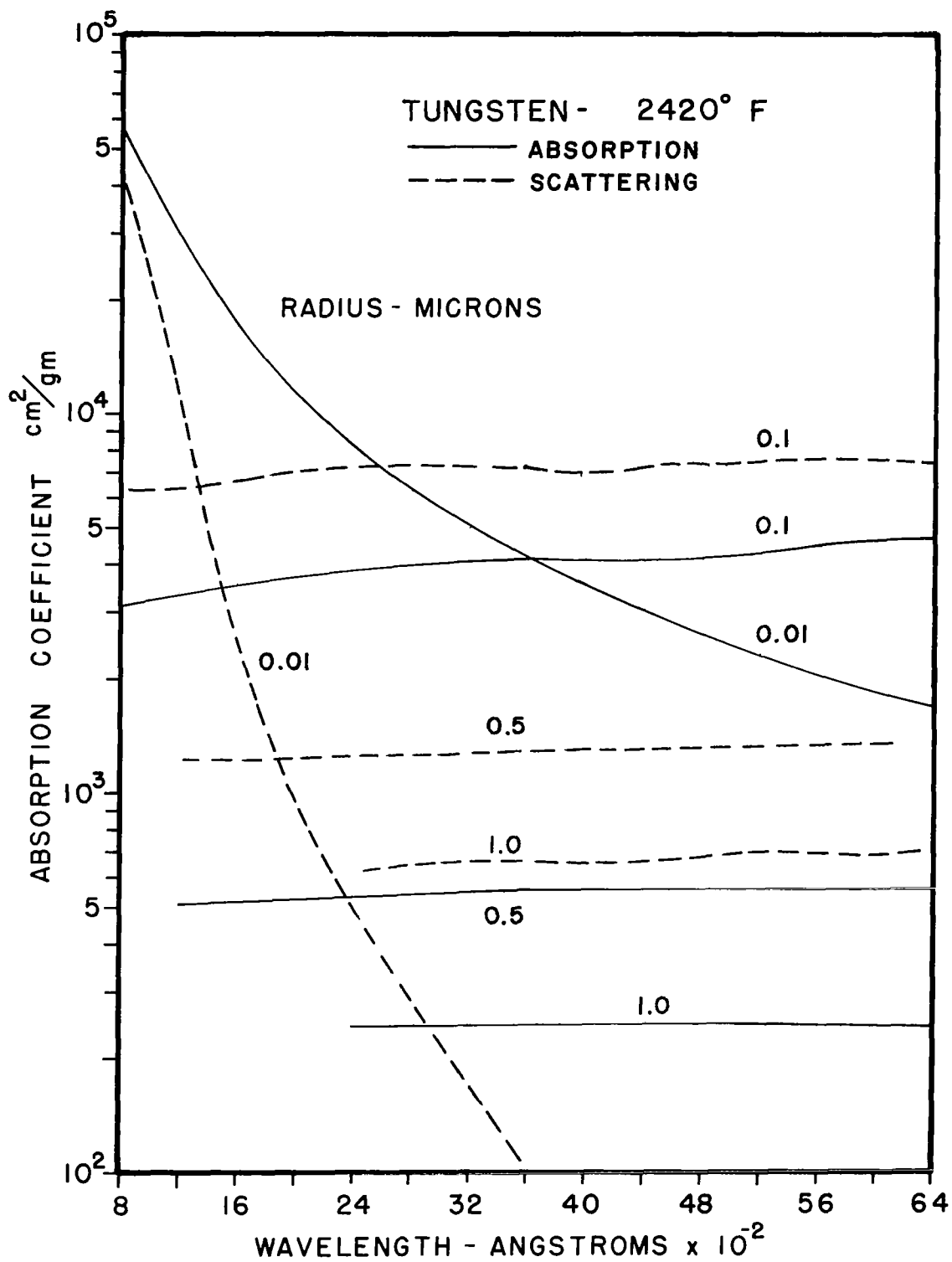
Figure 11. Theoretical Absorption and Scattering Parameters of Spherical Tungsten Particles at 76°F.

and scattering coefficients for spherical particles of tungsten do not change significantly with temperature. The scattering parameter is larger than the absorption parameter for particles of the order of 0.1 micron radius or larger. This means more energy would be removed from a beam by scattering than by absorption. Scattering may be neglected only if the particle radius is of the order of 0.01 micron. At these temperatures, for very small particle diameters, the absorption coefficient decreases with radiation wavelength. As in the case of carbon, the absorption coefficient for tungsten particles decreases as the particle size decreases. This effect is seen more clearly in Figures 12 and 13. At these temperatures, it can be seen that the absorption coefficient increases as the particle size decreases until a maximum value is reached and then decreases with particle size. Also, the value of the particle size at which the absorption is a maximum increases with radiation wavelength. Since the variation of the absorption coefficient with temperature is small, it is rather difficult to study the effect of temperature from Figures 10 through 13. In Table 1, the absorption coefficient of 0.2 micron radius tungsten particles is given at various temperatures and radiation wavelengths. From this table it can be seen that the temperature dependence is very small.

Figure 14 illustrates the extinction, absorption and scattering parameters of spherical particles of silicon at 80°F as a function of particle size for a radiation wavelength of 2400 Å. The absorption coefficient reaches a very high value for very small particle sizes.

In Figure 15, the absorption coefficients of spherical particles of three different materials are given for a radiation wavelength of 2000 Å as a function of particle radius. For particle radii greater than 0.1 micron carbon has the highest absorption coefficient, tungsten the smallest, and the absorption coefficient of silicon lies between the two. For particle radii smaller than 0.1 micron, silicon has the highest absorption coefficient compared to carbon and tungsten.

The theoretical values of the absorption parameter of carbon compare well with experimental values measured by passing a beam of radiant energy through a particle cloud suspended in hydrogen. For tungsten and silicon,
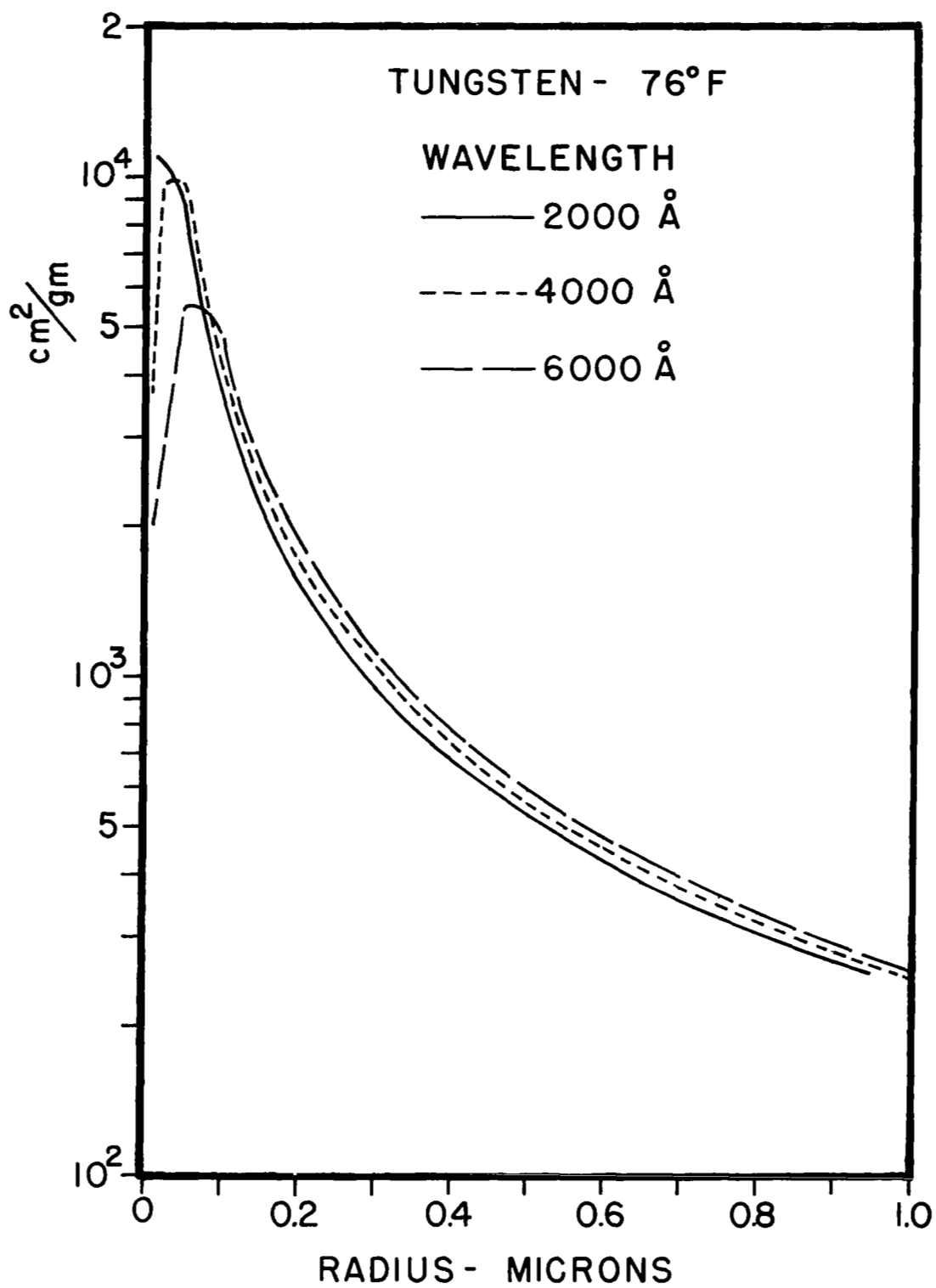
Figure 12.   Theoretical Absorption Parameter of Tungsten at 76°F
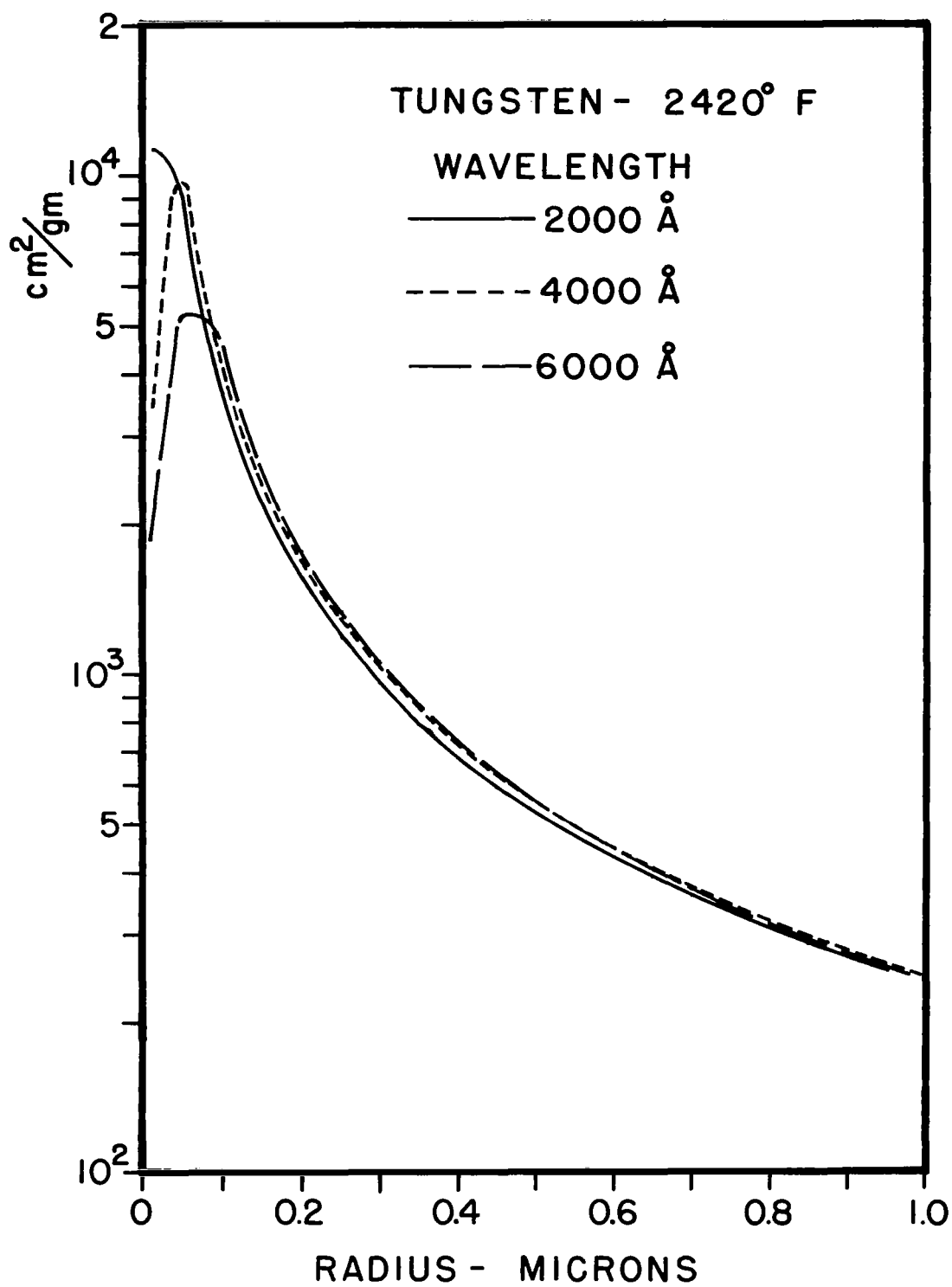as a Function of Particle Radius

Figure 13. Theoretical Absorption Parameter of Tungsten at 2420°F as a Function of Particle Radius

Table 1. Theoretical Absorption Parameter of 0.2 Micron Radius
Tungsten Particles at Various Wavelengths and Temperatures

| Wavelength	Absorption Parameter cm^2/gm		
(Å)	2420° F ($\times 10^4$)	1520° F ($\times 10^4$)	76° F ($\times 10^4$)
800	0.1377	0.1387	0.1365
1200	0.1456	0.1468	0.1442
1600	0.1505	0.1518	0.1493
2000	0.1544	0.1558	0.1534
2400	0.1587	0.1601	0.1583
2800	0.1613	0.1630	0.1618
3200	0.1642	0.1659	0.1655
3600	0.1676	0.1693	0.1694
4000	0.1692	0.1707	0.1707
4400	0.1733	0.1748	0.1743
4800	0.1738	0.1768	0.1762
5200	0.1719	0.1749	0.1760
5600	0.1743	0.1785	0.1815
6000	0.1755	0.1808	0.1859
6400	0.1795	0.1866	0.1939
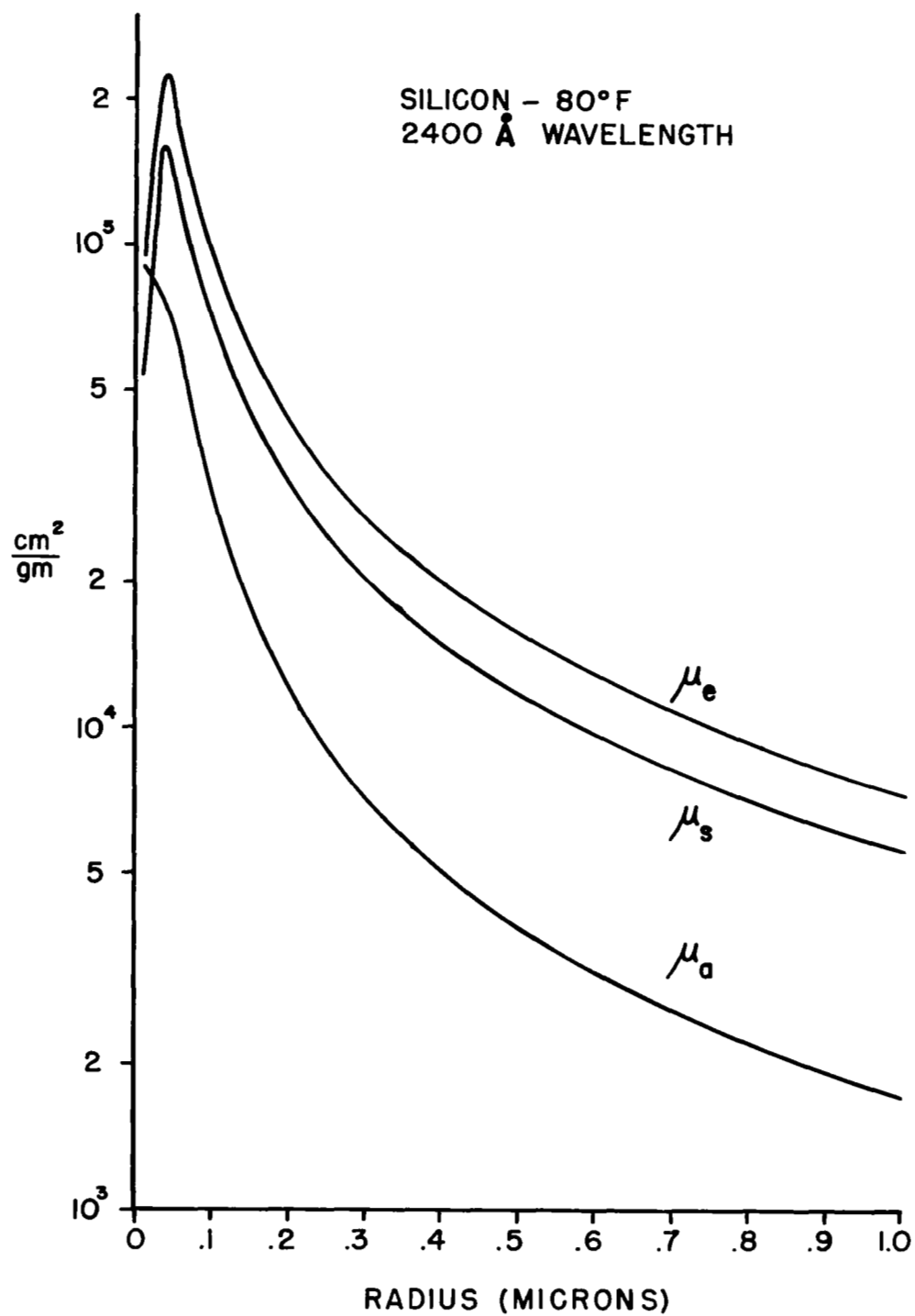
Figure 14. Theoretical Absorption, Scattering and Extinction Parameters
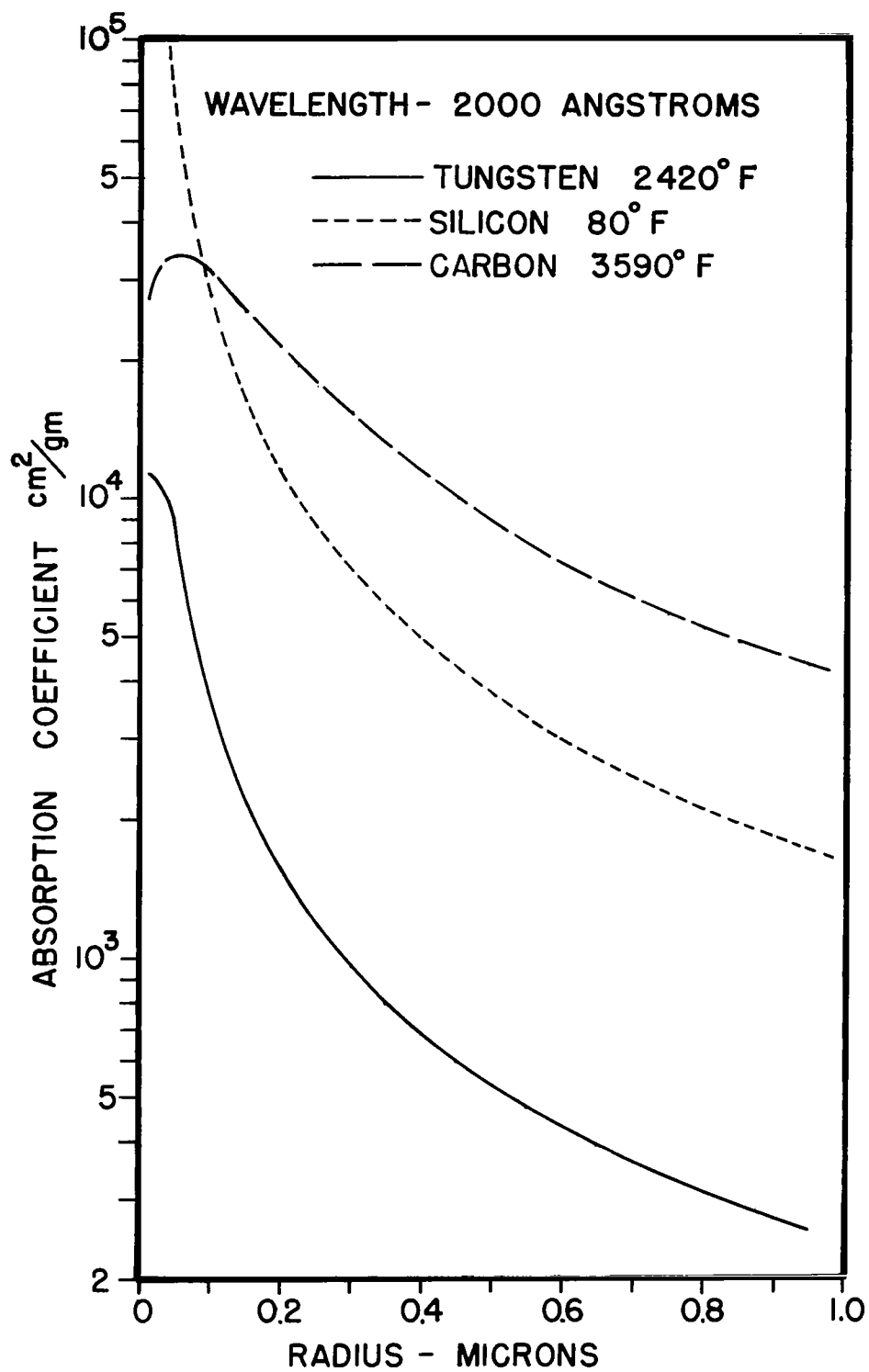of Spherical Silicon Particles.

Figure 15. Theoretical Absorption Parameters of Spherical Tungsten, Silicon and Carbon Particles.

however, the measured values are about a factor of two higher than the theoretical absorption parameters, probably due to the effect of scattering. This experimental program is described in detail in references 16 and 29. A more general description is provided by reference 30.

DISCUSSION OF RESULTS

It has been shown that in order to make calculations regarding radiant heat transfer to particle-seeded gases one must first evaluate the absorption and scattering characteristics of the seeded gas. These characteristics are specified by the following parameters: the linear absorption coefficient of the gas $k_a^g(\lambda)$, the mass density of particles in the aerosol ρ, the absorption parameter of the particles $\mu_a(\lambda)$, the scattering parameter of the particles $\mu_s(\lambda)$, and the scattering amplitude function of the particles $\rho(\lambda, \cos\theta)$, where θ is the angle of scattering. $k_a^g(\lambda)$ is a complex function of the gas properties as well as wavelength but may usually be evaluated for the pure gas. Values of $\mu_a(\lambda)$, $\mu_e(\lambda)$ and $\rho(\lambda, \cos\theta)$ may be calculated for spherical particles in a transparent medium using the Mie Theory, however, the Mie Theory serves only as an approximation when one is working with irregularly shaped particles. In general, $\mu_a(\lambda)$ and $\mu_s(\lambda)$ are not significantly temperature dependent over the range of temperatures investigated.

For particle radii greater than 0.1 microns carbon has a higher absorption parameter than silicon or tungsten. For particle radii smaller than 0.1 microns, silicon has the highest absorption parameter compared to carbon and tungsten.

CONCLUSIONS

1. Spherical carbon particles of about 0.2 microns diameter have a high absorption parameter of about 50,000 cm^2/gm and particles less than 0.1 microns diameter exhibit little scattering.

2. Tungsten particles exhibit a much lower absorption parameter than carbon. Tungsten particles of about 0.2 micron diameters have an extinction parameter of about 20,000 cm^2/gm and two thirds of that is scattering parameter, whereas carbon has an extinction parameter of about 50,000 cm^2/gm and is almost a pure absorber.

3. Silicon exhibits a higher extinction parameter ($\sim 65,000$ cm^2/gm) than either silicon or carbon, but more than two thirds of that is scattering parameter.

4. Although scattering itself does not heat the particle-seeded gas, scattering increases the average path length traversed by the thermal radiation and thereby enhances absorption. If the particles are small enough, about as much energy is back-scattered as is forward-scattered. In this case, scattering may significantly increase the absorption of thermal radiation.

5. Scattering is relatively unimportant for radiant heat transfer calculations to gases seeded with carbon particles of about 0.1 micron diameter or less, but may be very important if silicon or tungsten is used as the seed material.

6. The wavelength range of interest for gaseous reactor heat transfer calculations appears to lie between 2000 and 8000 Å.

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